Supporting Information

Coherent and Compact Zinc Electrodeposition Enabled by Compressing the Electric Double Layer of the Deposits

Ruirui Zhao[1], Haifeng Wang[1], Haoran Du[1], Ying Yang[1], Zhonghui Gao[1], Long Qie[1, 2*], and Yunhui Huang[2*]

[1] Institute of New Energy for Vehicles, School of Materials Science and Engineering, Tongji University, Shanghai, 201804, China. [2] State Key Laboratory of Material Processing and Die & Mold Technology, School of Materials Science and Engineering, Huazhong University of Science and Technology, Wuhan, Hubei Province, 430074, China.
*Corresponding author.
E-mail: qie@tongji.edu.cn; huangyh@hust.edu.cn

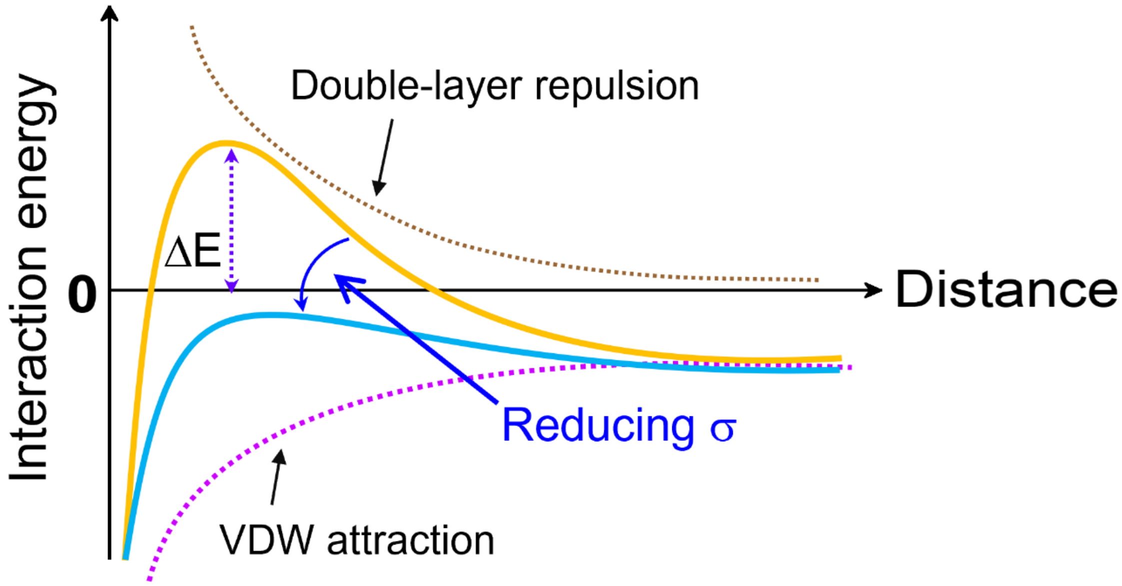

Figure S1 The schematic of DLVO interaction energy curves.

In a dilute aqueous solution, the forces between two particles are predicted based on the Derjaguin-Landau-Verwey-Overbeek (DLVO) theory[1-2]. According to the DLVO theory, a van der Waals (VDW) force and an electrostatic force act between two surfaces or particles. A VDW behaves attractively for two particles, while an electrostatic force behaves repulsively due to the same kind of charges. In this work, the electrostatic force for Zn deposits is the electrical double layer (EDL) repulsion force[3-4]. As shown in **Figure S1**, the VDW attraction is prominent when the particles possessing low charges and two particles attract mutually.

Otherwise, two particles repulse mutually. In conclusion, the EDL repulsion force depends on the net charges of the particles. The VDW attraction depends mainly on the distance between the particles[5].

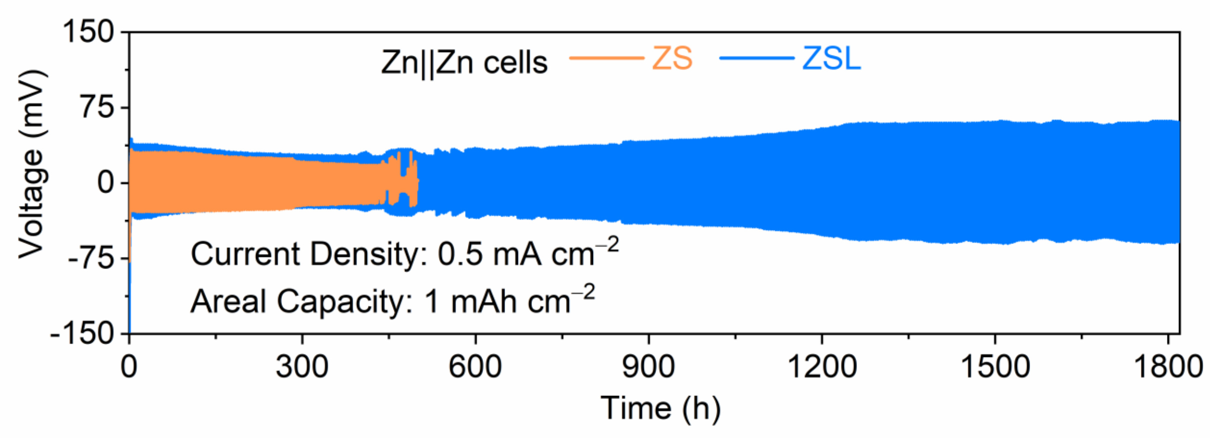

Figure S2 The plating/stripping performance for Zn electrodes in Zn||Zn symmetrical cells with ZS and ZSL electrolytes under a current density of 0.5 mA cm^{-2} for 2 h.

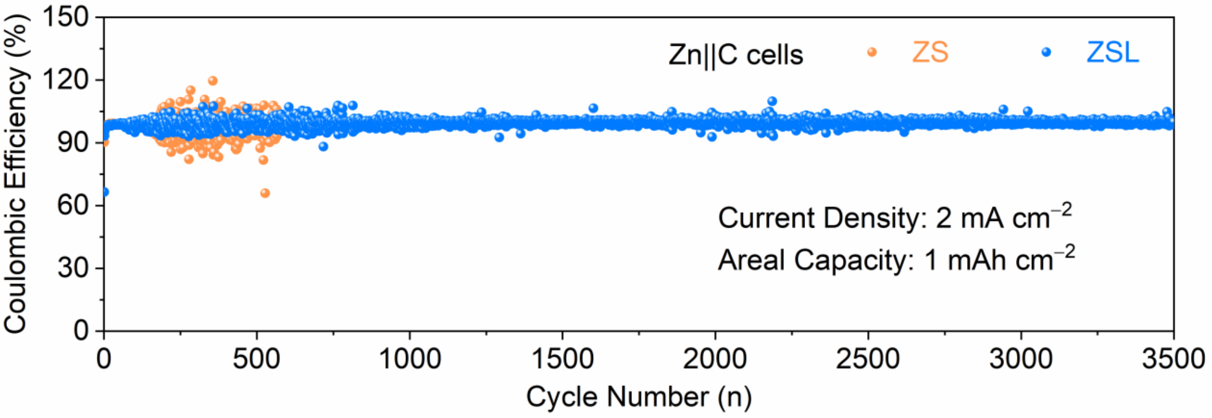

Figure S3 The CE of Zn||C cells with ZS and ZSL electrolytes a current density of 2 mA cm^{-2} and an areal capacity of 1 mAh cm^{-2}.

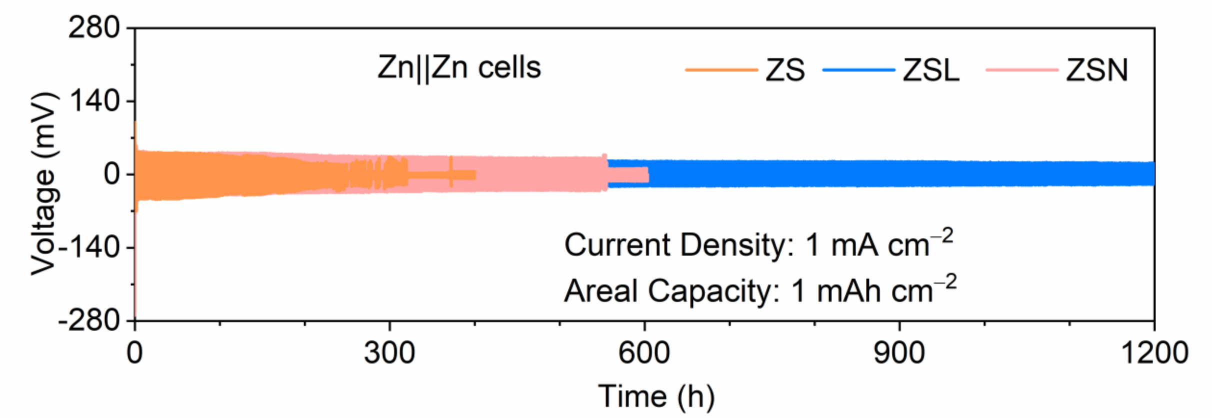

Figure S4 The Zn plating/stripping performance of Zn||Zn cells in ZS, ZSL, and ZSN electrolytes.

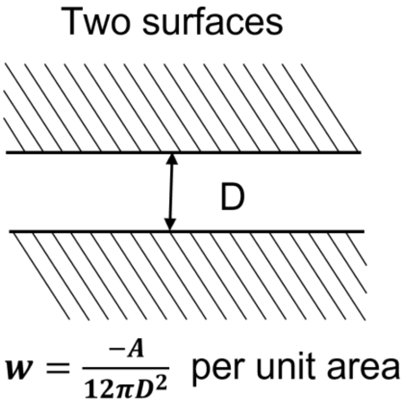

Figure S5 Van der Waals interaction free energies for two planar surfaces based on the Hamaker summation method.

In this work, the VDW free energy for Zn deposits is calculated according to the interaction forces between two surfaces. The Hamaker constant A is defined as the equation S(1):

$$A = \pi^2 C \rho_1 \rho_2 \qquad S(1)$$

where ρ_1 and ρ_2 are the number of atoms per unit volume for the surfaces, and C is the coefficient in the atom-atom pair potential. As a result, VDW attraction is largest when Zn deposits stack along the (002) plane of the closest packed plane crystal structure.

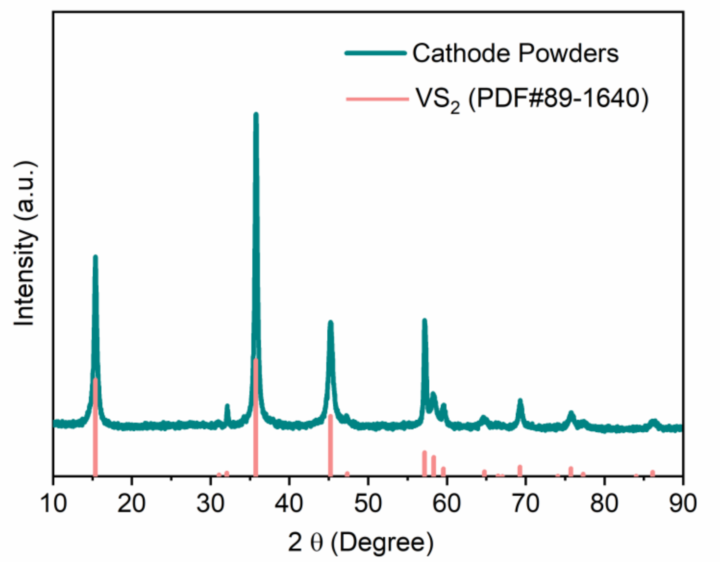

Figure S6 The XRD pattern of the VS_2 powder.

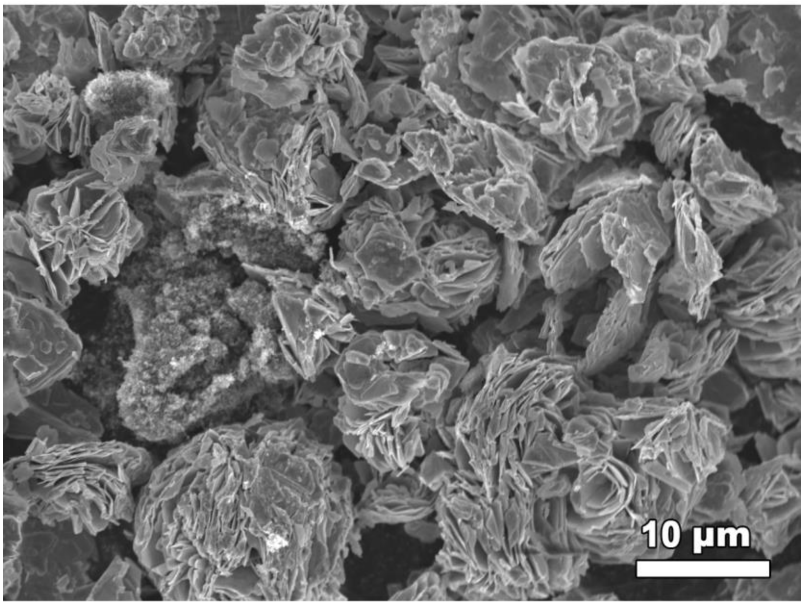

Figure S7 The SEM image of the VS_2 powder.

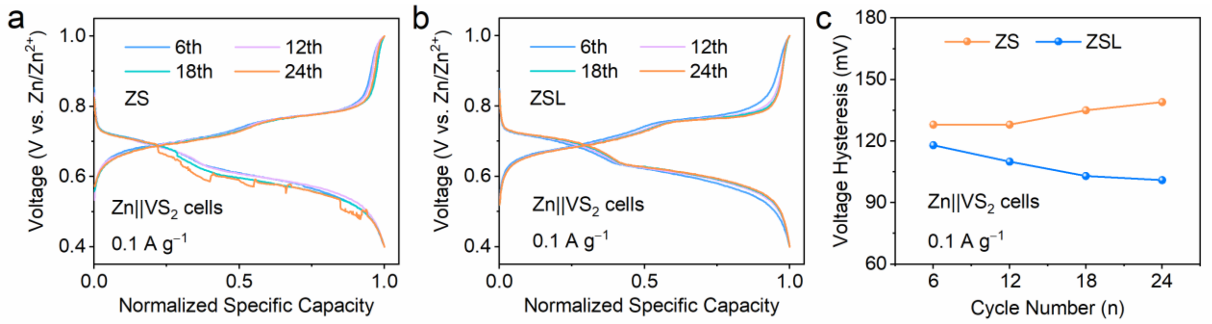

Figure S8 The normalized charge-discharge curves of Zn||VS$_2$ cells with a limited Zn supply in ZS (a) and ZSL (b) electrolytes at different cycles under a current density of 0.1 A g^{-1}; the comparisons of voltage hysteresis of Zn||VS$_2$ cells at medium discharge-capacity in both electrolytes (c).

Reference

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